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Autoreferát disertační práce
Summary of the Ph.D. Thesis

Tvorba a transformace atmosférického aerosolu v mezní vrstvě

Formation and transformation of atmospheric aerosol in boundary layer

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Abstrakt

Experimentální měření počtu aerosolových klastrů od velikosti 1,17 nm probíhalo od srpna 2016 do prosince 2018 na Národní atmosférické observatoři Košetice. Analyzovány byly podmínky atmosféry, které vedou ke stabilizaci klastrů, formování aerosolových částic a jejich následnému růstu. Během dní, kdy nedocházelo k formování částic, se projevil vliv nárůstu výšky mezní vrstvy atmosféry způsobující pokles koncentrací celkového počtu aerosolových částic i plynných polutantů. Naopak při událostech vzniku nových částic byl efekt zředění atmosféry překryt nárůstem počtu nových částic a byl pozorován pouze pokles koncentrací plynných polutantů. Mezní vrstva atmosféry byla během vzniku částic vyšší, což může znamenat obohacení atmosféry o další látky přenášené dálkovým transportem nebo jejich přenos z volné troposféry. Měření menších velikostí částic aerosolového spektra pomohlo lépe specifikovat dny a podmínky atmosféry, kdy nejenže dochází ke stabilizaci klastrů, ale nově vzniklé částice i nepřerušeně rostou.

Abstract

The experimental measurement of aerosol clusters from 1.17 nm in size was carried out from August 2016 till December 2018 at the National Atmospheric Observatory Košetice. Atmospheric conditions leading to aerosol clusters stabilization, fresh particles formation and particle growth were analyzed. Data of days with no new particle formation confirmed the connection between mixing layer height development and decrease of total aerosol number concentration together with lower gaseous pollutant concentrations. On the contrary, new particle formation process overcomes dilution of the atmosphere by increasing the number of freshly nucleated particles. Only decreasing gaseous pollutant concentrations were observed during these events. The atmospheric boundary layer was high during new particle formation events that can mean enrichment of the atmosphere by other components transported by long-range transport or some transfer from the free troposphere. The measurement in smaller sizes of the aerosol spectrum helped to better specify the days and conditions when stabilization of aerosol clusters occurred, and newly formed particles grew noninterruptedly.

1. Introduction

Atmospheric aerosols (AA) as solid or liquid particles suspended in the atmosphere affect the climatic system of planet Earth. Presence of AA in the atmosphere directly influences radiation balance by scattering and/or absorption of both solar radiation and longwave Earth radiation on aerosol particles. Indirectly, AA affect the radiation balance via interaction between radiation and clouds. AA serve as cloud condensation nuclei and thus affect clouds formation and further precipitation. Interaction with radiation is influenced by chemical composition, size, shape or concentration of AA (Boucher et al., 2013; Hinds, 1999; Pöschl, 2005). The number of particles directly emitted into the atmosphere can be estimated from anthropogenic sources, for example, using emission inventories. However, the number of secondary formed particles depends on the gaseous precursors' concentrations and atmospheric conditions (Dada et al., 2017).

Particles formation and their consequent growth are denoted as New Particle Formation (NPF) events. This phenomenon, involving both particle formation and their transformation to bigger sizes, is considered one of key parameters influencing the climate (Kulmala et al., 2004b; Dall'Osto et al., 2018; Pushpawela et al., 2018). Thus, scientific community has focused on NPF events studies. NPF events were observed in several geographical areas and different types of environments during the last decades

(Kulmala et al., 2004a; Pushpawela et al., 2018). Some studies stated that NPF events are suppressed in a polluted environment (Dada et al., 2017; Ling et al., 2019). Other studies report opposite results (Pikridas et al., 2015; Zhang et al., 2016; Tuovinen et al., 2020).

Parameters of the environment which contribute to particles formation in permanently changing atmosphere are still not fully clarified. State-of-art technologies enable the scientific community to perform experimental measurements in smaller and smaller aerosol spectrum sizes (Dada et al., 2018; Dong et al., 2019; Lampilahti et al., 2020). Simultaneous measurements of molecular cluster formation, meteorological conditions and chemical composition of the atmosphere head towards finding such relationships in the atmosphere, which undoubtedly lead to particle formation and their consequent growth as well. Atmospheric conditions affecting aerosol particles formation were investigated in this thesis by experimental state-of-art technologies at the background station National Atmospheric Observatory Košetice. The data on freshly nucleated aerosol clusters were analysed from 1.17 nm in size upwards.

2. Aims of the study

This thesis aims to:

To install and operate the unique experimental measurement of aerosol cluster concentrations in the Czech Republic.

To classify aerosol particles concentrations data and identify days when NPF events occurred.

To analyse atmospheric conditions (using data from air quality monitoring and meteorological data measurement) which favor or suppress molecular clusters formation and their consequent growth.

To develop a methodology for processing raw data on Atmospheric boundary layer height and using this data to evaluate the connection between Atmospheric boundary layer height and NPF events.

To analyse the linkage between Atmospheric boundary layer height and aerosol cluster formation.

To determine the possible dependence of location of local, regional and long-range sources on the particle growth rate.

3. Material and methods

The research for this study was conducted at National Atmospheric Observatory Košetice (NAOK, $49\,°34'24''$ N, $15\,°4'49''$ E, 534 m a. s. l.). NAOK is a rural background station at Czech-Moravian Highlands. The station owner is Czech Hydrometeorological Institute, and this station is both a part of National air quality monitoring network and a professional meteorological station. NAOK is also a core station of the Large research infrastructure ACTRIS-CZ and therefore is equipped with state-of-art technologies focused on advanced aerosol research.

This research used data from three main instruments - nano Condensation Nucleus Counter – nCNC (A11, Airmodus, Finland), Scanning mobility particle sizer - SMPS (custom-made, IfT Tropos, Germany), and Ceilometer (CL51, Vaisala, Finland). Counter nCNC allows to measure molecular cluster concentrations from 1.17 nm in size. The scanning mode and six size categories were used (1.17—1.26 nm, 1.26—1.44 nm, 1.44—1.76 nm, 1.76—2.34 nm, 2.34—3.41 nm, $>$ 3.41 nm). SMPS measurement range is 10—800 nm in 70 size categories. Ceilometer is an instrument aimed for Atmospheric boundary layer (ABL) height monitoring.

The auxiliary data from air quality measurement (concentrations of: sulphur dioxide - SO2, carbon monoxide - CO, ground-level ozone - O_3, nitrogen oxides - NO_X, particular matter - PM_{10} and $PM_{2.5}$, organic carbon - OC) and meteorological parameters (mixing layer hight - ML, atmospheric boundary layer hight - ABL, global radiation - GLB, air temperature - T, air pressure - P, relative humidity - RH, wind speed - WS, wind direction - WD) were used for detailed analyses.

All the data used in this work fulfil the quality requirements according to World Meteorological Organisation, international programme Aerosol, Clouds and Trace Gases Research Infrastructure and EU Directive 2008/50/ES (quality assurance are different by different groups of data).

Days of studied period (August 2016 – December 2018) were classified as day with:

- New Particle Formation - NPF
- No New Particle Formation - NON
- Undefined - UND event.

This classification was based on Particle Number Size Distribution - PNSD data from nCNC and SMPS instruments. Own classification was done for nCNC data, SMPS data was classified according to the method suggested by Dal Maso et al. (2005).

As descriptive characteristics for detailed analysis were calculated:

- Condensation sink (CS), determining the rate of molecule condensation (the loss of molecules) onto the pre-existing aerosol, was computed by integrating the PNSD (Kulmala et al., 2004a).

- Coagulation sink (Cc) is a parameter representing the rate of freshly nucleated particles coagulation onto pre-existing particles (Kulmala et al., 2012).

- Particle growth rate (GR) of particles freshly formed during NPF event days was calculated from the time evolution of Geometric mean diameter (GMD) values of particles smaller than 100 nm (Jeong et al., 2010).

- Formation rates (FR) of molecular clusters were calculated by Dal Maso et al. (2007).

- Proxy H_2SO_4 concentration was computed (Petäjä et al., 2009) since direct measurement of H_2SO_4 was not available in the Czech Republic. H_2SO_4 present in the gas phase in the atmosphere is considered the crucial factor for NPF (Zhang et al., 2012).

The potential location of sources influencing selected variables was computed in two ways:

- The local or regional transport was estimated from the relation between wind speed, wind direction, and a selected variable, obtained by Conditional Probability Function (CPF). The CPF polar plots, illustrating the probability of occurrence of a concentration at the given wind direction and wind speed, were calculated by the R package Openair (Carslaw and Ropkins, 2012).

- Long-range transport was investigated through backward trajectories calculated by HYbrid Single-Particle Lagrangian Integrated Trajectory HYSPLIT_4 model (Draxler, Roland R. and Rolph, 2013). The backward trajectories were used both for the Potential Source Contribution Function (PSCF) calculated by TrajStat plugin in MeteoInfoMap software (Wang et al., 2009) and for air masses cluster analyses.

The own methodology of processing the Ceilometer data was developed based on Lotteraner and Piringer (2016) procedure. The two main parts of ABL were selected - the mixing and boundary layer for their heights processing. The original 16-second data were resampled to 5-minute and hourly intervals and daily cycles fitted with polynomial regression function (Cleveland et al., 1992).

4. Results and discussion

Selected characteristic of aerosol transformation process was compared to the height of ABL and ML. Differences between NON and NPF event days were also analysed for total aerosol number particle concentrations, cluster concentration in 5 size categories, Condensation sink, Coagulation sink and Formation rate.

4.1. Atmospheric boundary layer height and total aerosol particles number concentrations

The ABL reached an average height in the range 1.5 to 2 km during the studied period. ML reacted on increasing solar radiation intensity by developing its height from morning hours with culmination after 3 PM UTC. ABL and ML pronounce a degree of atmosphere dilution - low ML means low dilution level of the atmosphere, high ML means conditions suitable for atmosphere dilution.

The decreased total aerosol particles number concentrations from morning hours caused by atmosphere dilution connected with ML height development was observed during NON event days (minimal concentration $2.5 \cdot 10^3$ $\#\cdot\mathrm{cm}^{-3}$ maximum ML height of 1200 m). On the contrary, the diluting effect of the atmosphere during the daytime was overcome by increasing total aerosol particle number concentration because of particle formation on NPF event days ($6.8 \cdot 10^3$ #·cm^{-3} at ML height 1400 m).

4.2. Cluster concentrations, Condensation and Coagulation sink, Formation rate

Molecular cluster concentrations increase was more pronounced in the smallest size category (1.17–1.26 nm) compared to rest of size categories, especially during NON events. The rise of molecular cluster concentrations in all size categories occurred during NPF events (starting from $0.4 \cdot 10^2$ #·cm^{-3}), the concentration increased 3–5 times). Dilution of the atmosphere probably does not affect the smallest size category's molecular cluster concentrations (observed at NON and NPF events). However, atmosphere dilution's influence on lowering cluster concentrations in the rest of size categories during NON events was not excluded.

CS and Cc values depend on pre-existing particles concentration in the atmosphere. Their daily evolution more or less reacted on ML height development. The increase of ML height generally means decreasing CS and Cc values caused by lowering pre-existing particles concentrations by the atmosphere dilution. CS daily amplitude during NON events (2.70–$3.34 \cdot 10^{-2}$ s^{-1}) did not fluctuate as much as CS during NPF events (2.40–$3.43 \cdot 10^{-2}$ s^{-1}).

The formation rate of the molecular clusters in all size categories showed similar daily evolution as cluster concentration. The highest FR was observed at size category 1.17–1.26 nm. Concentrations raised from 1.20 to 1.60

$cm^{-3}{\cdot}s^{-1}$(NON events) and from 1.10 to 5.04 $cm^{-3}{\cdot}s^{-1}$ (NPF events).

4.3. Meteorological conditions during NON and NPF events

Atmospheric conditions during NON and NPF events were compared to determine what conditions are suitable for aerosol particle formation. Time series of selected characteristics (meteorological and air quality parameters) during NON and NPF events were tested by Wilcox-Mann-Whitney test (Bauer 1972; Hollander et al. 2013). Statistically significant differences between individual pairs of the atmospheric components on the significance level of 0.05 were proved. Detected differences NON versus NPF events were:

- Gradually decreasing ABL height from a maximum at 6 AM UTC.
- The lower temperature by 2–5 °C.
- The lower maximum of global radiation 400 $W{\cdot}m^{-2}$ (NPF 700 $W{\cdot}m^{-2}$)
- The lower air pressure
- Higher relative humidity by 15%
- Prevailing West and Southeast wind (NPF West and Northwest wind)

- Higher concentrations of CO, NO_X, PM_{10}, OC
- Lower concentrations of SO_2 and O_3
- Lower cluster concentrations
- The higher CS
- The lower FR
- The lower proxy H_2SO_4 concentrations

4.4. Differences between classifications of NPF events

More detailed analyses were done for cases when the classification results of NPF event were different by nCNC and SMPS classification. From 426 days, 92 were classified as NPF event by both classification, 55 days were classified differently. Agreement in NPF classification (ANPFc) means that molecular cluster formation led to particle formation and further growth. Disagreement in NPF classification (DNPFc) results in particle formation without further growth. Circumstances that interrupted particle growth were analysed. Daily evolution of meteorological parameters was similar during ANPFc and DNPFc. Higher global radiation values, air temperature and air pressure were detected during ANPFc. From 2 to 8 AM, ABL and ML height was lower at ANPFc compared to DNPFc (Figure 4.1).

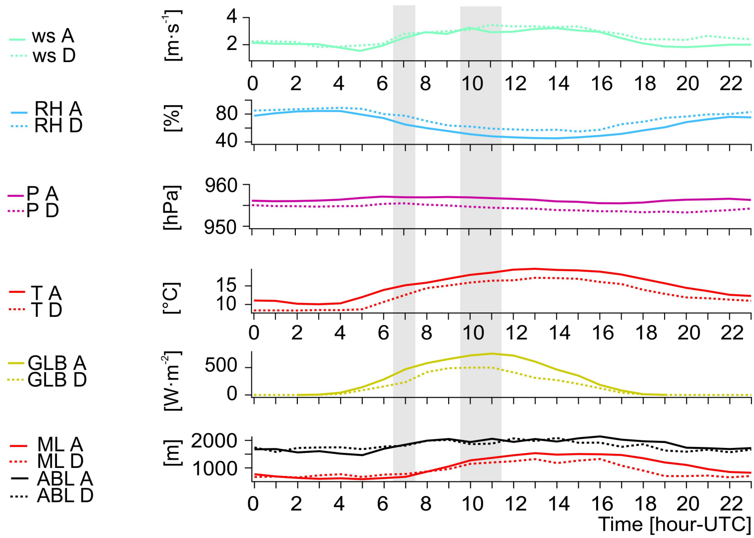

Figure 4.1: Daily cycle of meteorological parameters in case of agreement (A) and disagreement (D) in classifications. Gray bars highlight the beginning of the increase in cluster concentration and the time of number clusters concentration maximum during the agreement in the classifications. The solid lines show the agreement, dotted line disagreement. ML - mixing layer, ABL - atmosphere boundary layer height, - global radiation, T - temperature, pressure - atmospheric pressure, RH - relative humidity, ws - wind speed.

Differences in atmospheric components behavior were visible in SO_2 and H_2SO_4 concentrations between ANPFc and DNPFc. SO_2 concentration started to decrease at the same time when cluster formation begins and H_2SO_4 concentrations culminated during ANPFc (7 AM UTC). On the contrary during DNPFc, SO_2 concentrations were unstable and H_2SO_4 increased from morning hours and reached its maximum at 11 AM (Figure 4.2).

Atmospheric conditions in the first part of the day (0–12 AM UTC) seem to be a crucial for NPF. Time series divided into two groups (0–6 and 6–12 AM) during ANPFc and DNPFc were tested by Wilcox-Mann-Whitney test. Statistically significant differences (significance level of 0.05) were proved between meteorological parameters except between ML and ABL in time interval 6–12 AM, and between WS in both time intervals. Statistically significant differences on the significance level of 0.05 were detected by SO_2 and O_3 concentration and for CS by both time intervals. Results of Wilcox-Mann-Whitney test proved statistically significant differences by cluster concentration 1.17–1.27 nm, FR and proxy H_2SO_4 concentrations in time interval 6–12 AM (Figure 4.3).

These results indicate that noninterupted particle growth is effected by global radiation values, air temperature, air pressure and relative humidity and concentrations of SO_2 and O_3. The air masses origin influence to NPF events were analysed by clustering of 1739 backward trajectories. Six mean air clusters were calculated – clusters number 1–4 have some maritime history, clusters number 5 and 6 are continental (Figure 4.4).

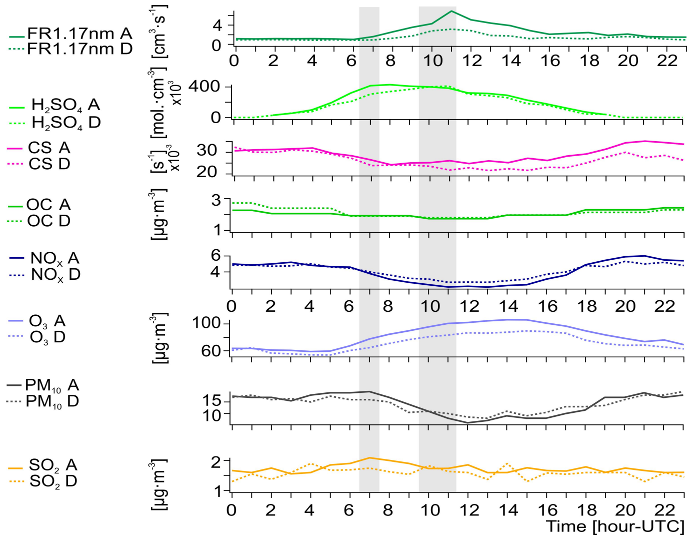

Figure 4.2: Daily cycle of pollutant concentrations, H_2SO_4 proxy and rate growth of clusters. Gray bars highlight the beginning of the increase in cluster concentration and the time of number clusters concentration maximum during the agreement in the classifications. The solid lines show the agreement (A), dotted line disagreement (D). SO_2 - sulfur dioxide, PM_{10} - particles up to 10 μm, O_3 - ground-level ozone, NO_X - nitrogen oxides, OC - organic carbon, CS - condensation losses, H_2SO_4 - sulfuric acid, FR - cluster formation rate.

Statistically significant differences between selected parameters in air mass clusters during ANPFc and DNPFc were tested by Dunn test with Holm method (Dinno, 2015;

Dunn, 1961).

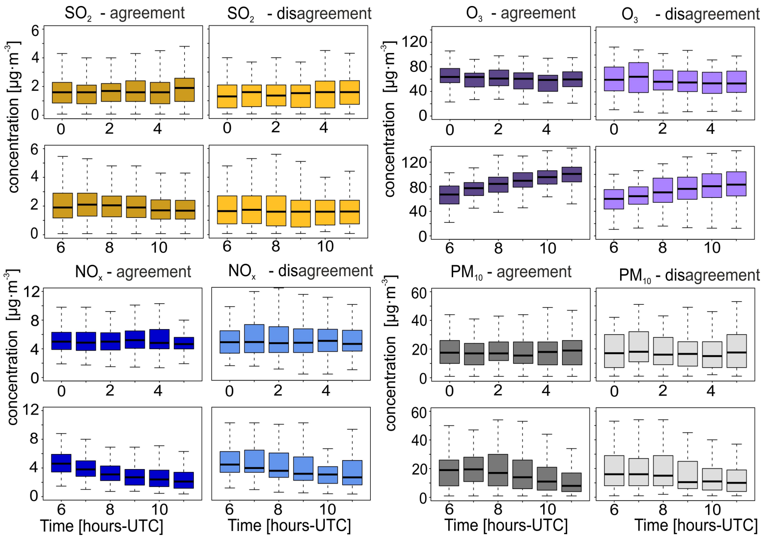

Figure 4.3: Box-whiskers graphs of the pollutant concentrations evolution between 0–6 and 6–12 hours, for days when the nCNC and SMPS classifications were in agreement and disagreement. SO_2 - sulfur dioxide, PM_{10} - particles up to 10 μm, O_3 - ground-level ozone, NO_X - nitrogen oxides.

Results did not proved that Air mass origin influenced molecular cluster formation in size 1.17–1.26 nm, ML and ABL height, or H_2SO_4 concentrations. However, Air mass origin affects Total number concentrations (Air mass cluster no. 1–4), O_3 concentrations (Air mass cluster no. 1,

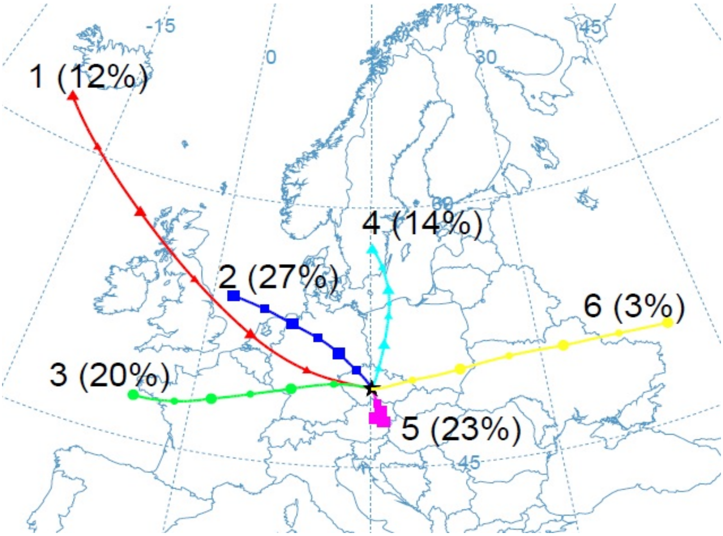

Figure 4.4: Cluster analysis of air mass backward trajectories. The position of the receptor is indicated by a black star.

2, 5), and CS values (Air mass cluster no. 1, 4, 5).

4.5. Growth rate of aerosol particles in different types of environment

Highly time-resolved PNSD were evaluated during five years (2013–2017) at four background stations in the Czech Republic located in different types of environments - agricultural background (National Atmospheric Observatory Košetice), urban background (Ústí nad Labem), industrial background (Lom), and suburban background (Prague-Suchdol). The PNSD data was used for new particle formation events determination as well as growth rate (GR) and condensation sink (CS) calculations.

Condensation sink reflects the pollution load at the individual station and its connection to the environment type. The response of CS to the atmospheric boundary layer (ABL) evolution was most visible at NAOK. CS dropped ($9.0 \cdot 10^{-3}$ s^{-1}) after sunrise and kept its low values ($7.5\cdot$ 10^{-3} s^{-1}) until the evening hours . At the urban and suburban stations Ústí n/L and Suchdol, the connection with morning and evening traffic rush hours was quite pronounced. In these two cases, increasing CS values after 3 AM peaking at 7 AM (CS–$1.7\cdot$ 10^{-2} s^{-1} and $1.2\cdot$ 10^{-2} s^{-1}) were observed. The secondary maximum observed at all stations between 9–12 PM was a result of the decreasing boundary layer height. At Lom, a different CS daily pattern was found. The decrease of values after morning peak continued only slowly, and a secondary maximum at noon was recorded (Figure 4.5).

The median growth rate (4 $nm\cdot h^{-1}$) was very similar at all stations, and corresponds well to the results of similar stations reported in recent studies (Bousiotis et al., 2019; Kerminen et al., 2018; Nieminen et al., 2018). The most frequent start time of the growth was from 10:00 to 12:00 UTC across the stations. The most typical length of growth was 2–4 hours at all stations. The second highest frequency was recorded in length category 0–2 hours, except for NAOK station with the 4–6 hours being more frequent. A unique GR length time lasting more than 10 hours with frequency over 10% was recorded at NAOK (Figure 4.6).

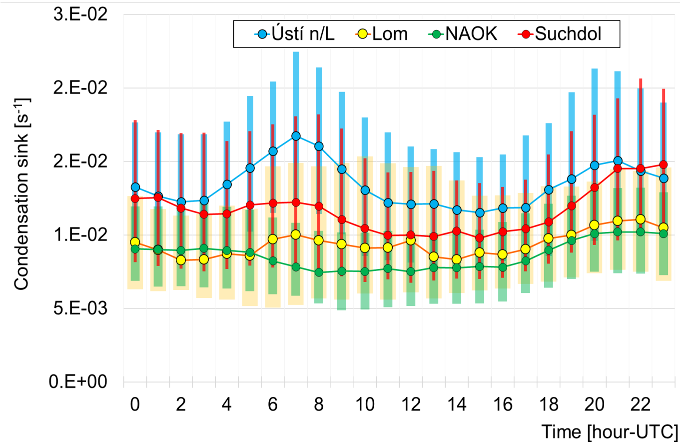

Figure 4.5: The median daily cycle of CS at stations the Ústí n/L, Lom, NAOK, and Suchdol (denoted by markers). The top/bottom border of the boxes represents the 75th and 25th percentile, respectively.

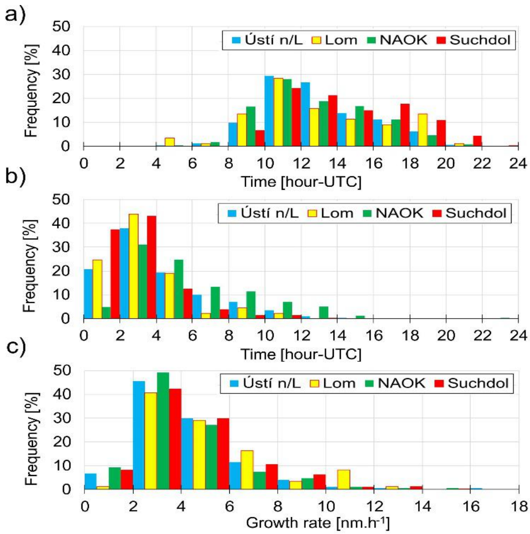

Figure 4.6: GR characteristics during NPF events at Ústí n/L, Lom, NAOK, and Suchdol. a) frequency of the start time of the particle growth, b) frequency of the length of the particle growth, c) frequency of the growth rate.

Within the space of the Czech Republic, GR was influenced by SW, SE and E sources. The east part of the Czech Republic is strongly influenced by heavy industry. Industrial complexes specialized in coal and steel processing are located close to the Czech-Polish border. Other industrial regions in the Czech Republic did not influence the GR values at the studied stations (Figure 4.7).

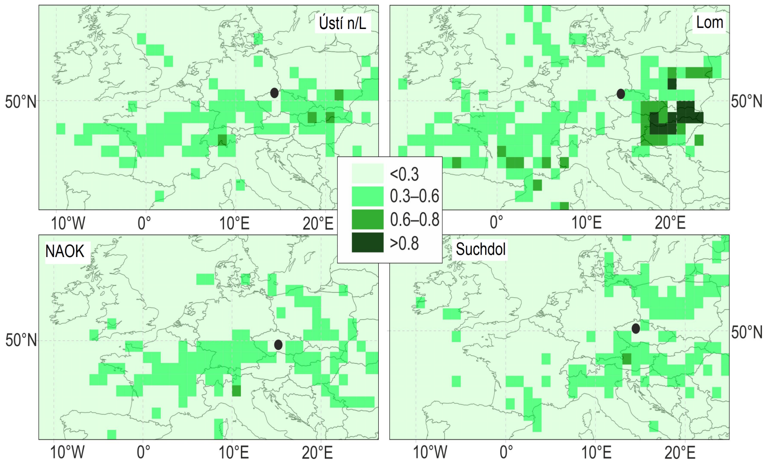

Figure 4.7: PSCF of the sources calculated for threshold set to 75$^{\text{th}}$ percentile of GR values. The black point indicates the station's position - Ústí n/L, Lom, NAO Košetice and Suchdol.

5. Conclusions

This study focuses on aerosol particles formation in Atmospheric boundary layer. Markers for transformation process were New Particle Formation events. The following results emerged from the overall data analysis:

- From 426 days of measurement, 119 were classified as NPF event days (27.5%) and 172 as NON event days (40.4%). This ratio is consistent with previous results or with similar type of station (Zíková and Ždímal, 2013; Nieminen et al., 2018).
- Molecular cluster concentrations in size range 1.17–3.41 nm were approximately 5× lower during NON events compared to NPF event
- Sufficient molecular cluster stabilization to growth by condensation processes were indicated by higher concentration in size category 2.34–3.41 nm compared to concentrations in categories 1.44–1.76 nm and 1.76–2.34 nm.
- NON events CS values were 1.2× higher between 6 AM to 1 PM UTC compared to NPF event days. It probably indicates suppressing conditions for NPF.
- Daily evolution of FR were 2× higher on average during NPF events.

- Wilcoxon-Mann-Whitney test proved statistical significant differences between meteorological parameters during NON and NPF events. Values of global radiation were 2.5× higher on average during NPF events.

- Dilution effect caused by ML height increase from morning hours was observed during NON events (Total aerosol particle number concentrations decreased). On the contrary, formation of new particles overcome dilution effect during NPF events.

- Only SO_2, O_3, and proxy H_2SO_4 concentrations were higher during NPF events.

- The analysis of differences in NPF event classification by nCNC a SMPS shows that particle formation without growth interruption was affected by atmospheric conditions in time period 0–12 AM UTC. Statistically significant differences between ANPFc and DNPFc were proved in global radiation, air temperature, air pressure, relative humidity, SO_2, O_3 concentrations, and CS values.

- Parameter OX that indicates photochemical activity was higher after sunrise during ANPFc.

- The highest frequency of statistically significant differences was found between characteristic of air masses number 4 (maritime), 1 (fresh maritime) and 5 (aged continental). Origin and history of air masses did not directly affect molecular cluster concentrations, SO_2 and NO_X concentrations, and ML and ABL height. However, O_3 concentrations were influenced by air mass origin.

- Receptor location is probably influenced by long-range transport of SO_2.

- GR at Czech background station was on average $4\,\mathrm{nm \cdot h^{-1}}$. Influence to GR from local sources proved effect of transport, industry and heating. Effect of Long-range transport to GR from industrial European region was not proved.

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Curriculum vitae

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Education:

2016–present	Charles University Faculty of Science Doctoral programme of study Field of study: *Environmental studies*
2007–9th July 2009	Masaryk University Faculty of Science Master's degree program Field of study: *Physical Geography*
2004–2007	Masaryk University Faculty of Science Bachelor's degree program Field of study: *Physical Geography*

Courses:

2019	3rd Conference and training schoool: *Time series analysis* Tromsø, Norway
2018	2nd HAAR Summer school Navarino Environmental Observatory, Greece
2017	EUFAR STANCO Training Course Cambridge, UK
2014	27th GAWtec Zugspitze, Germany
2013	9th Summer School on Atmospheric Aerosol Physics Measurement and Sampling Hyytiälä, Finland

Work Experience:

2020–present	Czech Hydrometeorological Institute Košetice Observatory *Head of the Košetice Observatory*
2020–present	Czech Hydrometeorological Institute Košetice Observatory Project ACTRIS-CZ *Junior Researcher*
2014–2019	Czech Hydrometeorological Institute Košetice Observatory *Researcher*
2013–2019	Global Change Research Institute of the CAS *Technician*
2011–2014	Czech Hydrometeorological Institute Košetice Observatory *Sampling Manager*

2010–2011	Valeo Compressor Europe s.r.o. Humpolec *Quality Laboratory Technician*

Skills:

Computer programs:	Basic knowledge: R - Open air, IgorPro User knowledge: ArcGis, CorelDRAW, MS Office

Selected publications:

Adéla Holubová Šmejkalová, Naděžda Zíková, Vladimír Ždímal, Helena Plachá and Miroslav Bitter. Atmospheric aerosol growth rates at different background station types. *Environmental Science and Pollution Research.* 2020. doi: 10.1007/s11356-020-11424-5.

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